INÍCIO

GEOCHEMISTRY OF MINERALS, WATERS AND WEATHERING FROM THE FONTE SANTA MINE AREA (NE OF PORTUGAL)

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Abstract

The quartz veins containing scheelite from Fonte Santa mine area were exploited for W between 1942 and 1982. At the end of November 2006, a flood event damaged the dam land of Fonte Santa mine and metal content of water increased. Fonte Santa mine area cuts the quartzites close to the Fonte Santa muscovite granite. The granite contains quartz, microcline, albite, muscovite, chlorite, columbite-tantalite, volframite, W-ixiolite and ilmenite. The quartz veins contain muscovite, chlorite, tourmaline, scheelite, pyrrhotite, pyrite, sphalerite, chalcopyrite, galena, arsenopyrite, magnetite, jarosite, phosphates of Pb, Fe and Al. The waters related to the Fonte Santa mine are poorly mineralized, with electrical conductivity < 965 µS / cm, of mixed type or HCO_3^- and SO_4^{2-} types. These waters have Fe and Mn contents that forbid to use that for human consumption and agriculture. Sodium, Mg and K water contents are associated with the alteration of albite, chlorite and muscovite of country rock, while Ca is related to the W-bearing quartz veins.

Key-words: mineralizations, scheelite, waters, contamination

Resumo

Os filões de quartzo contendo scheelite da mina de Fonte Santa foram explorados para W entre 1942 e 1982. No final de Novembro de 2006, uma inundação destruiu a barragem de terra das minas da Fonte Santa e o teor de metais na água aumentou. A mina da Fonte Santa corta os quartzitos, próximo do granito moscovítico de Fonte Santa. O granito possui quartzo, microclina, albite, moscovite, clorite, columbite-tantalite, volframite, W-ixiolite e ilmenite. Os filões de quartzo possuem muscovite, clorite, turmalina, scheelite, pirrotite, pirite, esfalerite, calcopirite, galena, arsenopirite, magnetite, jarosite e fosfatos de Pb, Fe e Al. As águas relacionadas com a mina da Fonte Santa são pouco mineralizadas, com condutividade eléctrica <965 µS / cm, de tipo misto ou HCO_3^- e SO_4^{2-}. Nestas águas os teores de Fe e Mn impossibilitam a sua utilização para consumo humano e agricultura. Os teores de Na, Mg e K associam-se à alteração de albite, clorite e muscovite das rochas regionais e os de Ca aos filões de quartzo com tungsténio.

Key-words: mineralizações, scheelite, águas, contaminação

1. Introduction

Mining activities were very important for the Portuguese economy. About ninety abandoned mining areas show distinct contamination levels, but only a few of them were submitted to environmental recovery. However, Neves Corvo and Panasqueira mines are

still active and have an international dimension. The abandoned mining sites are frequently located close to occupied rural areas and some of the waters and soils are used for agriculture or human consumption without any assessment of environmental and human health risks (Abreu *et al.,* 2008).

Sulphides are stable and very insoluble under reducing conditions, but oxidation takes place when minerals are exposed to atmospheric conditions. The weathering of sulphide minerals promotes the formation of sulphuric acid, together with ferrous and ferric sulphates and ferric hydroxides, which lead to acidic conditions on the environment (Bell, 1998). The water pollution by dissolved metals in mining areas has mainly been associated with the oxidation of sulphide-bearing minerals, producing acid waters and a high level of dissolved metals (Cánovas *et al,.* 2008; Navarro *et al.,* 2008). The extent and degree of heavy metal contamination around mines vary depending upon geochemical characteristics and degree of tailing mineralizations. The metals released by sulphide oxidation are attenuated by precipitation, co-precipitation and sorption reactions (Berger *et al.,* 2000) in the mines and around them. However, the content of elements in the environment also depends on their mobility and solubility from rocks and stream sediments to waters.

Mining and mine waste release heavy metals into the environment, often due to the unsatisfactory disposal of waste rocks, causing problems. When water infiltrates into the waste rocks, the oxidation of sulphide minerals, specially pyrite, and their subsequent dissolution, can result in acid mine water and elements may be carried out to waters. Effluents of abandoned mine workings typically consist of acid mine drainage, eroded material from mine tailings and waste rocks.

The aim of this paper is to present a study of the geochemistry of minerals from scheelite quartz veins and associated waters from the area, comparing waters from inside and outside Fonte Santa mining area. The geochemistry of minerals from granites and W-bearing quartz veins were used to model the water-rock interaction process that raise the geochemistry of waters supplied in the studied area.

2. Geological setting

The Fonte Santa area is located in the South border of the Mirândes Plateaux, Northeast of Trás-os-Montes, included in the autochtonous Central Iberian Zone (ZCI) (Fig.1). The climate is characterized by an average annual temperature of 12.3 °C, with a very hot summer, with temperatures reaching 40.2 °C and cold winters with temperatures going down to -12.6 ºC. The water samples were collected during 2007, that was an atypical meteorological year (SNIRH, 2009).

The tungsten exploitation started in 1941 and the maximum production of mine was reached in 1953 and the exploitation stopped in 1982. The main quartz veins were exploited in open pits and underground.

The mine country rocks consist mainly of Lower Ordovician chlorite phyllite with rare intercalations of Armorican quartzites and Cambrian metasediments with magnesian marbles which crop out close to the area (Silva, 2000).

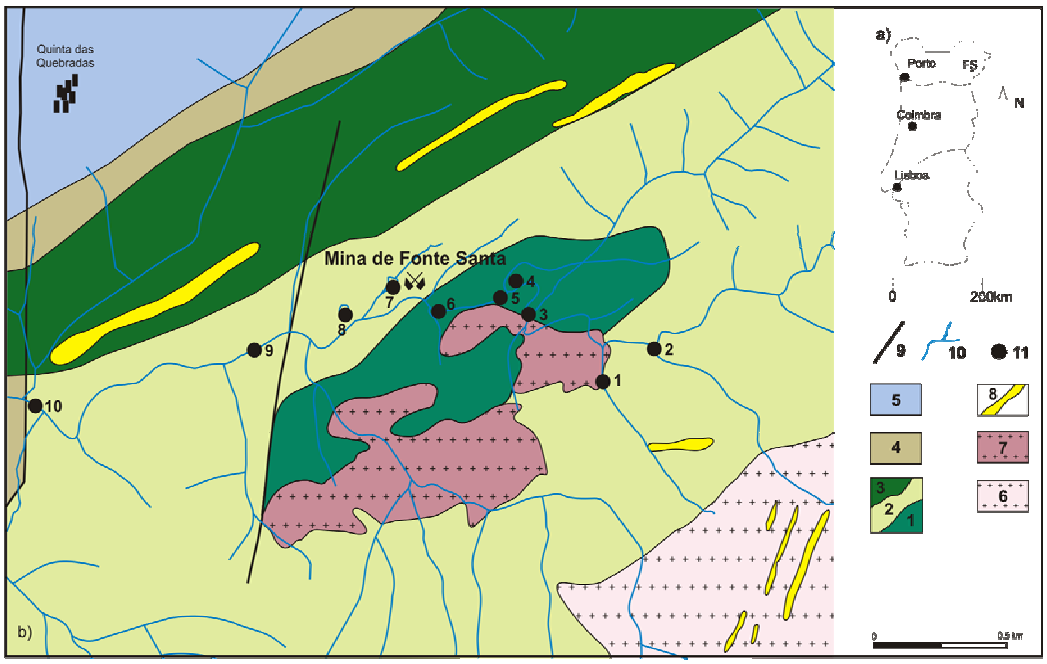

Figure 1 – a) Location of Fonte Santa mine area (FS), northeast Portugal; b) Geological map of the Fonte Santa mine (modified from Silva, 2000). Ordovician Quartzitic Marão Formation: 1. Lower quartzites, 2. intermediate schists, 3. upper quartzites, Ordovician schists-Moncorvo Formation: 4. slates, 5. lower Silurian quartzites, 6. medium- to coarse-grained porphyritic muscovite-biotite granite (G1), 7. fine- to medium-grained muscovite granite (G2), 8. quartz veins, 9. faults, 10. drainage lines, 11. sample points.

The mine is associated with a strong structural control, the Bemposta-Moncorvo shear zone, and is emplaced along the opened tension cracks (Parra *et al.*, 2001). The mineralization was controlled by litological and structural factors. The oldest is an irregular lenticular quartz vein, folded by the third Variscan deformation phase and the most recent one forms a *stockwork* with shafts orientated along the tension and shear cracks. The scheelite mineralization occurs mainly in quartz veins at pelitic levels and is also in skarns due to granite metasomatism on magnesian marbles and lies in the apical area of Fonte Santa muscovite granite massif, in the core of an antiform. The mineralized area is 300 m wide and 1100 m long, elongated along the ENE-WSW direction parallel to the regional structures. The maximum depth exploited is about 200 m and the material volume is about 20 million cubic meters (Ribeiro and Rebelo, 1971). Alluvionar scheelite occurs in the bead stream and on the alluvionar lands.

3. Analytical techniques

Minerals from the muscovite granite and associated quartz veins containing scheelite were analyzed on a Cameca Camebax electron microprobe and on a Jeol JXA-8500F at LNEG, S. Mamede de Infesta, (Portugal). Analyses were carried out using an accelerating voltage of 15 kV and a beam current of 20 nA and 10 nA were used in the Cameca Camebax and the Jeol microprobes, respectively. Each element was counted for 20 seconds. Beam diameter was 5 μm for most analyses except for mica where a beam of 10 μm was used. Standards used include albite (Na Kα); orthoclase (Al Kα, Si Kα; K Kα); cassiterite (Sn Lα); $MnTiO_3$ (Mn Kα, Ti Kα); Fe_2O_3 (Fe Kα); sphalerite (Zn Kα, S Kα); pyrite (S Kα, Fe Kα); galena (Pb Mα); wollastonite (Ca Kα, Si Kα); AsGa (As Lα); MgO (Mg Kα); Au (Mα); Mo (Lα); Ni (Kα); Cu (Kα); As (Lα); Ag (Lα); Co (Kα); Cd (Lα); Sb (Lα); Bi (Mα); Ta (Mα); Nb (Lα); Mo (Lα) and W (Lα, Mβ).

The Fonte Santa exploitation mining area and the location of water samples are presented in Fig. 1. Ten water sampling points were selected and water was collected at each point four times a year (January 2007; April 2007; August 2007 and December 2007), obtaining a total of 38 water samples, because one water point (FS1) had no water in August and December due to Fonte Santa's dry climate. January and April of 2007 are considered as the wet season, whereas August and December of 2007 are the dry season.

Temperature, pH, Eh, electrical conductivity and alkalinity were determined *in situ*. The waters were filtrated, acidified and kept at 4 ºC. Anions were determined by ion chromatography with a Dionex ICS 3000 Model and cations were obtained by ICP-OES (Optical Emission Spectroscopy – Inductively Coupled Plasm) using a Horiba Jovin Hyvon JY 2000-2 Model. Arsenic was determined by flame atomic absorption spectrometry. The detection limit was 0.02 mg/L for most elements, except for As and K of 0.01 mg/L. The precision for most analyses was less than 5 %, but less than 15 % for Na (10.0 %) and Al (15.4 %). The laboratory analyses were performed at the Department of Earth Sciences, University of Coimbra (Portugal).

4. W-bearing quartz veins

The W-bearing quartz veins contain quartz, scheelite, sphalerite, galena, pyrite, pyrrhotite, chalcopyrite, arsenopyrite and rare siderite. Scheelite is anhedral, light coloured, with typical blue fluorescence. Either stolzite and ferritungstite fill fractures in scheelite. Wolframite was not found in quartz veins, but is a common acessory mineral in hydrothermally altered muscovite granite.

Quartz is partly recrystallized, locally brecciated and impregnated by different sulphides (Table 1) and fills many joints, fractures and microfissures.

Table 1. Average electron-microprobe analyses of some sulphides from W-bearing quartz veins of Fonte Santa mine.

	S	Zn	As	Fe	Mn	Ni	Cu	Cd	Pb	Ag	Sb	Bi	Total	n
Phyrrotite	39.33	0.42	0.04	59.58	_	_	0.07	_	_	_	_	_	99.44	7
Arsenopyrite	20.05	0.04	45.79	34.16	_	_	0.02	_	_	_	0.03	_	100.06	4
Sphalerite	32.60	57.00	_	10.00	0.35	_	_	0.54	_	_	_	_	100.49	8
Chalcopyrite	33.32	1.88	_	32.75	_	_	30.35	_	_	0.06	0.05		99.03	6
Pyrite	53.22	0.18	0.43	46.79	0.01	0.06	0.06	_	_	0.02	0.06	0.10	100.92	15
Galena	13.50	0.18	0.03	0.09	0.05	_	0.08	0.10	81.85	1.14	0.07	2.44	99.52	25

Elements in wt. %, - not detected, n- number of analyses

Monoclinic pyrrhotite is associated with chalcopyrite and occurs included in sphalerite. Some pyrrhotite crystals are replaced by pyrite. Sphalerite is one of the most abundant sulphide minerals and shows "chalcopyrite disease". Many exsolutions of sphalerite show ortogonal alignments (one direction parallel with chalcopyrite and phyrrotite and another one perpendicular with galena in fractures). Sphalerite contains Fe, Mn e Cd that can substitute Zn, but the total of them is below 12 wt.%. In general, each sphalerite grain has a homogeneous composition. Chalcopyrite was only found as blebs in sphalerite and is homogeneous, but has some silver up to 0.19 wt.% replacing Fe. Pyrite crystals are euhedral or subhedral and fractured. They have inclusions of galena and electrum. Pyrite locally fills fractures in quartz. Arsenopyrite is rare and has a composition of FeAsS.

Galena is abundant and occurs filling fractures in sphalerite or in subhedral crystals locally well developed showing fractures and margins replaced by Pb sulphate veins and replacing pyrite. Galena has some Bi, Ag and Zn.

Magnetite occurs in subhedral crystals and also associated with ilmenite, chlorite, Al, Fe, and Pb hydrated phosphates and Fe sulphates in brecciated fragments of quartz veins surrounded by quartz.

5. Geochemistry of waters

The results of chemical composition of waters from Fonte Santa mining area are presented in Table 2. To obtain reference background data from waters located outside the mine influence, samples of a spring (FS1) and a natural stream (FS2) were collected and analyzed. Natural stream waters (FS5, FS6, FS9, FS10) and mine lagoons (FS3, FS4, FS7, FS8) were located inside the mining area and receiving the influence of abandoned mining activities and mineralized veins, but only the stream waters were affected by the flood event. Water mine lagoons FS3 and FS4 receive waters from coarse-grained tailings, while the FS7 and FS8 mine lagoons receive waters from fine-grained tailings and rejected mining materials.

Table 2. Variations intervals for selected elements and parameters of waters from Fonte Santa mining area.

	pH	HCO_3^- (mg/L)	SO_4^{2-} (mg/L)	NO_3^- (mg/L)	Na (mg/L)	K (mg/L)	Ca (mg/L)	Mg (mg/L)	Fe (µg/L)	Mn (µg/L)	As (µg/L)
FS 1	5.7-5.9	9.0-11.7	3.4-4.3	7.8-10.1	3.9-4.1	3.2-3.4	3.0-3.1	1.1-1.2	38-40	43-45	bd
FS 2	6.6-7.9	10.8-32.1	1.8-2.5	0.1-1.2	2.7-7.5	0.6-2.7	1.7-4.9	1.0-2.5	41-236	4-40.8	bd-2.4
FS 3	5.0-8.0	17.5-30.1	3.2-5.9	0.1-4.5	4.8-7.5	1.0-2.9	3.2-4.1	1.2-2.4	129-196	13-49	bd-16.6
FS 4	6.4-8.5	15.7-31.6	3.2-5.9	0.1-16.3	4.6-8.0	1.1-2.9	2.9-4.2	1.2-2.4	116-203	7.0-49	bd-14.1
FS 5	6.2-7.9	13.5-21.3	5.5-64.3	0.2-2.5	4.7-7.5	1.1-3.1	4.2-12.9	2.5-7.2	191-5651	122-995	6.1-51.7
FS 6	5.9-7.8	10.8-23.1	2.6-28.5	0.6-1.5	3.7-14.8	1.6-2.8	1.6-5.2	0.1-3.2	70-1396	36-776	5.1-50.7
FS 7	3.4-3.5	-	192.8-423.4	<1.0	11.3-14.7	6.6-10.8	33.7-50.7	33.2-48.4	1399-3325	5706-9260	1.1-5.77
FS 8	5.2-6.3	7.0-15.7	91.6-172.3	< 1.2	6.2-13.1	4.5-7.6	18.4-25.0	14.9-24.3	158-270	787-928	7.4-51.5
FS 9	5.8-8.2	8.3-15.5	3.7-120.4	0.2-1.6	4.0-9.5	2.7-3.5	8.8-20.8	0.7-15.9	23-3523	50-481	7.4-31.3
FS 10	6.4-7.8	9.7-17.5	11.5-89.2	0.1-2.2	4.2-9.1	2.4-3.3	3.3-14.7	1.7-10.8	190-2284	79-153	5.6-51.1

- not found; bd - below detection limit.

Most waters from Fonte Santa area do not contain a dominant cation composition and plot in the mixed water type. Relatively to the anions, HCO_3^- and SO_4^{2-} water types dominate.

The waters from Fonte Santa plot mainly in the field of near-neutral/low metal waters, according to the classification of Ficklin *et al*. (1992). Most waters from the Fonte Santa mine area are poorly mineralized, but waters inside the mine influence are richer in metal contents than those from outside that influence, showing the effect of abandoned old mining activities on water quality. The mine lagoon water samples (FS7 and FS8, Table 2) have acid/high metal concentrations and also tend to have the highest Eh values, electrical conductivity, SO_4^{2-}, K, Ca, Mg, Mn, Al, Sr, Li and most metal contents, particularly water sample FS7. In Fonte Santa, there is no significant acid mine drainage, because the area contains a small amount of sulphides and scheelite quartz veins cut the regional schist and quartzite and carbonates contributed to the neutralization of the waters and promote the decrease of trace element contents.

The hydrothermal minerals of Fe-tungstite, stolzite, Fe-sulphates and Fe, Pb and Al phosphates found in the scheelite quartz veins could retain some metallic elements on their structure and consequently they are not present in significant concentrations on the waters. Secondary sulphate minerals play an important role in acid drainage and metal sequestration in surface environments (Hammarstrom *et al*., 2005).

The waters with the highest SO_4^{2-} and metal concentrations are associated with the most contaminated mine lagoons with the lowest pH values that receive water from fine tailings and rejected mining materials (Fig. 1, Table 2). This correlation can be associated with oxidation and dissolution of Fonte Santa sulphide minerals, such as pyrite, chalcopyrite, sphalerite, galena, arsenopyrite and pyrrhotite. Most element contents from water mine lagoons (FS3 and FS4) are similar or lower than those found on stream waters (Table 2), because these points receive water from coarse-grained tailings. Arsenic has an irregular distribution in Fonte Santa waters and in some water samples is below the detection limit (Table 2) which can be attributed to the rare occurrence of arsenopyrite or due to the possible precipitation of this element on stream sediments and soils. Metals such as Fe form oxyhydroxide compounds in aqueous medium which are able to complex with As compounds and precipitate out of the solution and decrease As contents from waters (Serfor-Armah *et al.,* 2006).

Although most major and trace element contents of metals in waters from Fonte Santa are low (Table 2), some of them exceed the accepted values for human consumption and/or agricultural use (Portuguese Law, 2001; 2007). The water from mine lagoon, FS7, is the most contaminated of the area and must not be used for human consumption. Some Fe and Mn water contents in FS5 to FS10 points are higher than parametric values defined for human consumption. Some of these waters must not also be used for agriculture due to their Fe and Mn contents. Most waters from the Fonte Santa area have NO_2^- (0.1 mg/L) content, above those recommended for human potable water. The environmental impact of this abandoned mine is not very high and the contamination problems are essentially related to the flood event that carried contaminant load by stream along 2 km from the tailings and with the mine lagoons FS7 and FS8.

6. Weathering and Hydrogeochemistry

The foregoing analysis discusses the assessment of atmospheric, anthropogenic and natural contributions of major cations and anions to the average composition of water (Figure 2).

The atmospheric inputs were deduced from the $[Cl^-]/[Y]$ ratios in seawater (Appelo and Postma, 2005) and the anthropogenic inputs from the $[NO_3^-]/[Y]$ ratios in fertilizers (Pacheco *et al*., 1999), assuming that all chloride and nitrate are derived from those sources.

Notwithstanding the correction of the total concentrations for the atmospheric plus anthropogenic inputs, the sulphate concentrations remain very high suggesting that, in parallel with the carbonic acid derived from CO_2 dissolved in soil water, a weak sulphuric acid derived from pyrite oxidation will also play a role as weathering agent.

The natural contributions to water composition are associated with weathering of albite, chlorite and muscovite from the local granites and metasediments, producing Na, Mg and K and weathering of sulphides and scheelite present in quartz veins, producing Ca. The weathering of rock-forming minerals and mineralizations will also result in precipitation of secondary phases such as clay minerals (halloysite, smectite, vermiculite), metal oxides (ferritungstite) and Fe sulphates. Independently from the specific weathering reactions, equivalent proportions of bicarbonate and/or sulphate will be released with the cations, depending on whether carbonic acid or/and sulphuric acid is/are the weathering agent(s). In Fonte Santa area, weathering of minerals promoted by the attack of carbonic acid to the crystal lattices represents 61% of the total weathering, meaning that CO_2 is the dominating agent.

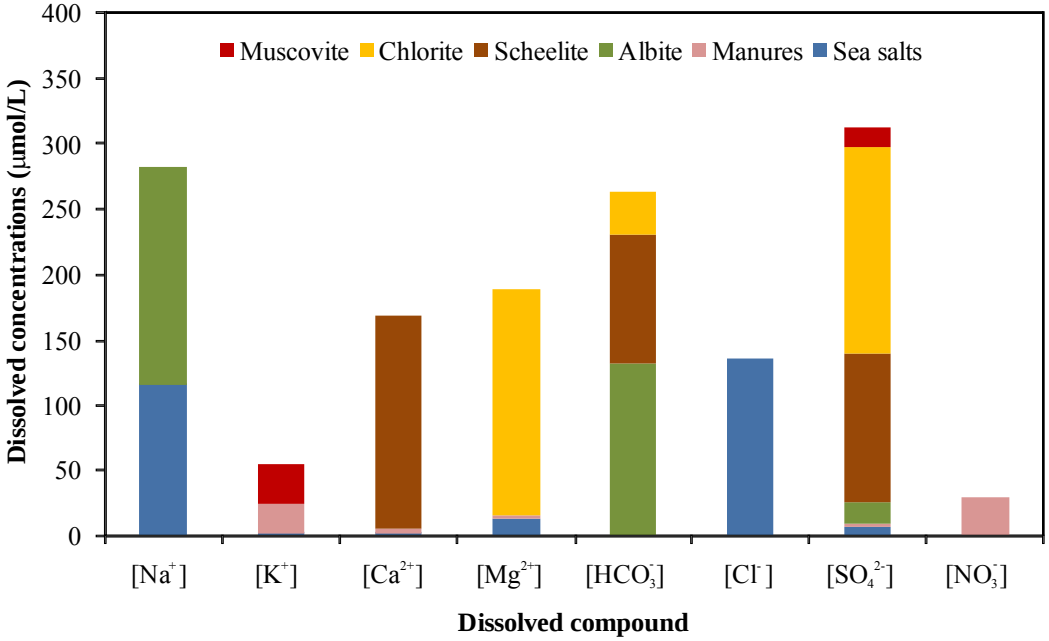

Figure 2 – Atmospheric, anthropogenic and natural contributions to water composition .

7. Conclusions

Mineral paragenesis of W-bearing quartz veins consists of scheelite, pyrite, pyrrhotite, sphalerite, chalcopyrite, arsenopyrite, galena, iron oxides, Al, Fe and Pb hydrated phosphates and Fe sulphates. Scheelite has fractures filled by stolzite and ferritungstite.

Waters from the Fonte Santa area are poorly mineralized. However, there is an increase in most parameters and element contents from outside to inside mine influence, showing the effect of abandoned old mining activities on water quality. Most of the waters from Fonte Santa do not contain a dominant cation-anion composition and are of mixed water type. Some of them are Na and Mg water types and HCO_3^- and SO_4^{2-} waters.

The environmental impact is essentially related to the flood event that carried contaminated load by stream increasing immediately Fe and Al contents in natural stream.There is no significant acid drainage associated with old mine workings, which can mainly be attributed to occurrence of calcium carbonates in country rocks that probably neutralized the waters and decrease metal concentrations.

Most waters associated with the mineralized veins and old mine activities at Fonte Santa have Fe and Mn concentrations that forbid their uses for human consumption and agriculture. Some waters show concentrations above parametric Portuguese values (such as SO_4^{2-}, NO_2^-, Mg, Zn, Al, Ni and Co) and should not be used for human consumption.

The alteration of albite, chlorite and muscovite of country rock are responsible for the sodium, magnesium and potassium present in water and the weathering of scheelite from W-bearing quartz veins is the most reasonable source for calcium. The weathering of rock-forming minerals and mineralizations will also result in precipitation of secondary

phases such as clay minerals (halloysite, smectite, vermiculite) metal oxides (ferritungstite) and Fe sulphates.

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